High-temperature high-pressure oscillating tube densimeter

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We describe an oscillating tube densimeter for use at temperatures up to at least 575 K and at pressures up to 20 MPa. After the densimeter was calibrated under vacuum and filled with water, it was used to measure the density of toluene from 298 to 575 K at 13.8 MPa. The results agree (0.1% rms deviation) with those obtained using other techniques. In the present densimeter, an alternating current is passed through the tube containing the sample to force the tube to oscillate in the field of a permanent magnet. This design avoids the use of electromagnets (with their attendant polymer or ceramic insulations) and does not require attachment of appendages to the oscillating tube. This densimeter oscillates in an overtone rather than in its fundamental mode, thereby achieving improved isolation from environmental noise and a shorter response time. The densimeter is small, weighing 370 g. If transformers are used to couple electrical signals to the oscillating tube, the densimeter itself may be constructed entirely out of metal.
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I. INTRODUCTION

Oscillating tube densimeters have been used to measure density of fluids accurately throughout a wide range of temperatures and pressures.[1–6] They require only small fluid samples that come into thermal equilibrium rapidly; thus, they are well suited to both on-line process control and to automated data acquisition in the research laboratory.

Here, we report our experiences with several novel design features intended to enlarge the range of temperatures and pressures accessible to oscillating tube densimeters as well as the time interval required between calibrations. One densimeter was used to measure the density of toluene in the temperature range 298 to about 575 K at a pressure of 14 MPa nine days after it was calibrated. The results had an rms fractional deviation of 0.09% from a published correlation.

The novel features discussed below include: (1) using overtone modes of oscillation for the determination of density, (2) driving the tube's oscillations by passing an alternating current through the tube itself while the tube is supported in the field of a permanent magnet, and (3) using transformers to couple the excitation and detection signals to the densimeter.

II. EIGENFREQUENCIES OF AN OSCILLATING TUBE DENSIMETER

For design purposes, it is useful to model the oscillating tube as a bar clamped at both ends. This model ignores the fact that the tube is bent into a U. In this crude approximation, the eigenfrequencies of the oscillations are determined by the boundary conditions that the displacement and the derivative of the displacement with respect to distance along the tube are both zero at the clamped ends. The eigenfrequencies are given by[7]

$$f_n=(\pi\kappa/2L^2)(Y/\rho_t)^{1/2}\beta_n^2. \tag{1}$$

Here, κ is the radius of gyration about the tube's axis, Y is Young's modulus, ρ_t is the mass density of the tube, L is the length of the tube, and β_n are eigenvalues. ($\beta_1=1.5056$, $\beta_2=2.4997$, and $\beta_n\simeq n+\frac{1}{2}$ for $n>2$.)

In analogy to a lumped constant oscillator, Eq. (1) can be rewritten as

$$2\pi f_n=(K/M_t)^{\frac{1}{2}}, \tag{2}$$

where K is an effective spring constant that is proportional to Y and M_t is the mass of the tube. When the tube is filled with liquid, the mass of the liquid is added to M_t in Eq. (2) to obtain

$$2\pi f_n=\left(\frac{K}{M_t+V\rho}\right)^{\frac{1}{2}}, \tag{3}$$

where the mass of liquid is expressed as the product of the liquid density ρ and the volume, V, within the tube. Rewriting Eq. (3) to express the density in terms of the resonance frequency, we have

$$\rho=(k/f_n^2-1)/v, \tag{4}$$

where $k(T)$ and $v(T,p)$ are temperature-dependent and pressure dependent functions to be determined through calibrations with two reference liquids of known density or with one liquid of known density (such as water) and with vacuum. In a highly idealized model, $k=K/4\pi^2M_t$ and $v=V/M_t$; however, straightforward generalizations of the model show that Eq. (4) applies even when the cross section of the tube is not uniform and even when the eigenfrequencies are not easily calculated because, for example, the tube is curved. In adopting Eq. (4), we retain the assumption that the fluid remains stationary within the tube and that the only restoring forces that contribute to the oscillation are the bending and shearing of the tube. Other phenomena, such as flowing of the liquid in and out of the tube during each cycle of the oscillation, are neglected.

We have chosen to operate the densimeter near the third (i.e., $n=3$) mode of oscillation. For a tube which is bent only

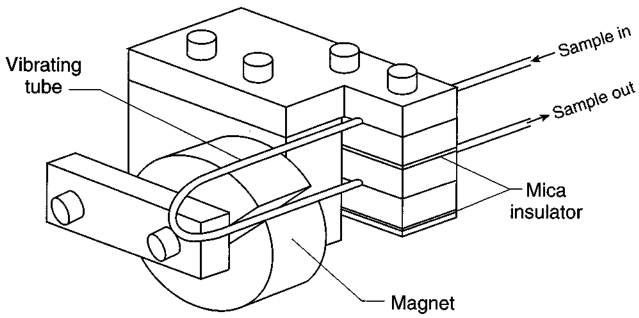

FIG. 1. Drawing of the densimeter.

a small fraction of a semicircle, this mode would be a nearly degenerate pair of modes with the resonance frequency for the in-plane mode being slightly lower than that for the out-of-plane mode. In the case of the U-shaped tubes that we studied, the frequency difference between the two modes ranged from 15 to 80 Hz for resonance frequencies near 4 kHz. (The quality factor Q of these modes was approximately 10,000.)

The in-plane $n=3$ mode has several advantages in comparison with the traditional choice, $n=1$. Because the frequency of the third mode is approximately five times the fundamental frequency, the present oscillator is much less susceptible to the low-frequency noise of the external environment, which tends to peak at low audio frequencies.

In the $n=3$ mode of oscillation, the two ''legs'' of the ''U'' bend towards and away from each other during the oscillation. To the extent that this motion is antisymmetrical, the center of mass does not move and the structure that supports the U does not recoil. This reduces the resonance frequency's dependence upon the method of support of the tube. (The effective mass of the oscillator is a reduced mass that accounts for the recoil of the supporting structure.)

Traditionally, the $n=1$ mode is used and the sensitivity of the oscillator to its support is reduced by supporting the tube on a massive base weighing several kilograms. The traditional approach requires thermostating the massive base. This requirement, in turn, increases the response time for temperature changes. In contrast, the densimeter shown in Fig. 1 weighed 370 g and was hung from a light frame by the thin, stainless-steel tab shown in Fig. 2.

III. EXCITATION AND DETECTION OF RESONANCE

The tube of the densimeter shown in Figs. 1 and 2 was forced to oscillate in the field of the permanent magnet by passing an alternating electrical current through the tube itself. In effect, the tube functions as a single-turn moving coil such as the coils that drive loudspeakers.

The present arrangement differs from many other densimeters that have permanent magnets fastened to the oscillating tube and are driven by nearby electromagnets that are exposed to the same environment as the tube itself. The present arrangement simplifies the mechanical construction of the densimeter, although at the cost of requiring more elaborate electronic instrumentation. We believed that simplifying the construction would result in a densimeter that was more stable, especially at high temperatures.

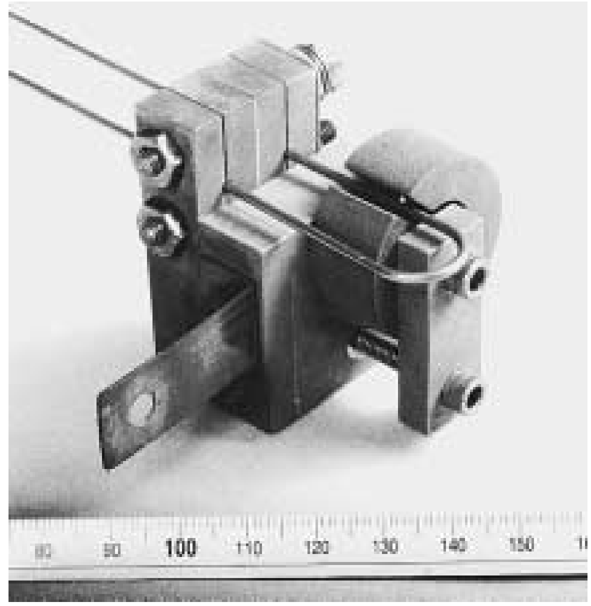

FIG. 2. Photograph of a densimeter. This densimeter was suspended inside a furnace by the tab fastened to the chair-shaped base plate in the front. In the prototype shown, the magnetic field was oriented 45° to the plane of the tube to study both in-plane and out-of-plane oscillations. The 90° orientation is recommended.

The present design eliminates the use of polymer or ceramic insulation used in electromagnets and it eliminates the use of a ceramic cement or a mechanical clamp to fasten the permanent magnet to the most sensitive element of the densimeter; namely, the oscillating tube.

Simple models of the excitation and detection schemes follow. The simplest circuit indicating the electrical connections to the oscillating tube is shown in Fig. 3. A more elaborate circuit that is discussed below is shown in Fig. 4. Both schemes have been tested and analyzed.

In the simplest circuit, the tube with an electrical resistance R_t is connected by two leads to an oscillator which generates a voltage E_0 across its internal resistance R_0. The interaction of the resulting current I with the magnetic field leads to a force $\mathscr{F}$ on the tube given by

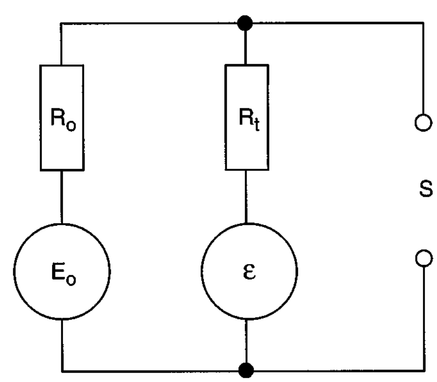

FIG. 3. Simple equivalent circuit for driving and detecting the motion of an oscillating tube densimeter with a current in the field of a permanent magnet.

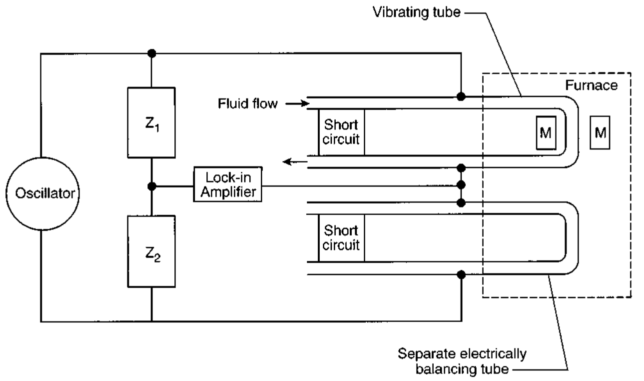

FIG. 4. Schematic of the bridge arrangement for the detection of the motional emf using a lock-in amplifier.

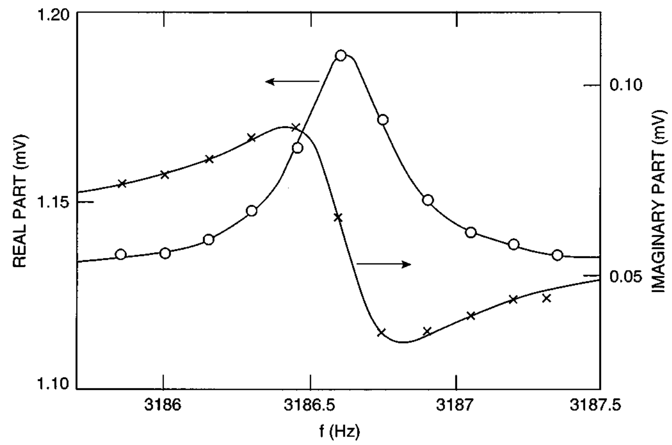

FIG. 5. A typical motional emf at the vicinity of a resonance. The signal was obtained without using the bridge arrangement. The complex signal is expressed in two parts: a real part (○), which is in phase with the driving emf, and an imaginary part (×) which is 90° out of phase with the driving emf.

$$\mathscr{F}=IB\ell. \tag{5}$$

Here B is the component of the magnetic field perpendicular to the tube and ℓ is the length of the tube in the magnetic field. The direction of the force is perpendicular to both I and B. When the tube moves perpendicular to the field with a velocity v, a motional emf with a magnitude ε given by

$$\varepsilon=vB\ell \tag{6}$$

is induced within the tube. The induced emf opposes the original emf E_0 that produced the current. Thus, the total current is given by

$$I=(E_0-\varepsilon)/(R_0+R_t) \tag{7}$$

and the signal voltage S at the output terminals (Fig. 3) is given by

$$S=IR_t+\varepsilon=\frac{R_t}{R_0+R_t}E_0+\frac{R_0}{R_0+R_t}\varepsilon. \tag{8}$$

Eq. (8) shows that the signal at the tube consists of two terms: one term is proportional to the driving emf, E_0, and the other to the motional emf, ε. The equation of motion for the coordinate y describing the oscillatory displacement of the tube is

$$M\frac{d^2y}{dt^2}+b\frac{dy}{dt}+Ky=\mathscr{F}, \tag{9}$$

where b is the mechanical loss coefficient that accounts for internal friction in the oscillating tube and for viscous damping of the tube's motion by the surrounding air. With a time dependence of $e^{i\omega t}$, the steady-state solution to Eq. (9) using Eqs. (5)–(8) is

$$S=\frac{R_t}{R_0+R_t}E_0\left[1+\frac{R_0 i\omega\gamma_m}{R_t i\omega(\gamma+\gamma_m)-(\omega^2-\omega_0^2)}\right], \tag{10}$$

where ω is the angular frequency of the driving emf and $\omega_0^2\equiv K/M, \gamma\equiv b/M$, and

$$\gamma_m=\frac{(B\ell)^2}{M(R_0+R_t)}. \tag{11}$$

The term γ_m is an additional loss term for the oscillator due to magnetic induction. When $\gamma\gg\gamma_m$, which is true for our case, the amplitude of the resonance signal is proportional to γ_m which in turn is proportional to B^2. Thus, maximizing B is an important design criterion.

For the densimeter shown in Fig. 1, $R_0=50\ \Omega$ and $R_t\simeq0.07\ \Omega$. At resonance, the value of the second term in the square brackets of Eq. (10) is only 5% of the constant term. Thus it is possible, but not straight forward, to use the oscillating tube as the frequency determining component in a feedback loop to sustain oscillations at the resonance frequency, and we have done so.

The alternative that we have followed is to use a frequency synthesizer as the electronic oscillator to drive the tube. Under computer control, the driving frequency was stepped through the resonance frequency and the detected signal S was measured with a lock-in amplifier and recorded. A typical example of the resulting data is displayed in Fig. 5.

Both the in-phase (○) and quadrature (×) components of S are plotted on an expanded scale with appropriate zero offsets. Also plotted is a six-parameter Lorentzian function [Eq. (10)] that has been fitted to the data. Two of the fitted parameters are the resonance frequency and the damping constant, two are a complex baseline, and two are a complex amplitude. For the data in Fig. 5, the resonance frequency and the damping parameters are 3186.650 Hz and 0.4375 Hz, respectively, and the quality factor is $Q=\omega_0/(\gamma+\gamma_m)=7283$. For an oscillator with such a high Q, the overtones have the additional advantage with respect to the fundamental mode that they may be scanned more quickly.

In circumstances where the reduction of the expense and complexity of the instrumentation is important, a bridge arrangement such as that shown in Fig. 4 is useful. If the bridge is balanced at a frequency that differs from the resonance frequency by a few times the resonance width, the motional emf at resonance can be readily detected. The bridge can be readily used as the frequency determining element of an oscillator.

We were able to obtain the large signal-to-noise ratio evident in Fig. 5 with a power dissipation of only 0.3 μW in the tube. Thus, the heating of the tube (and the sample) *via* the current used to drive the tube was entirely negligible. The total voltage drop across the tube (and the fluid in the densimeter) was about 1.5 mV, a voltage that is much too small to induce significant electrochemistry in the liquid under study.

The electrical conductivity of the fluid in the densimeter is in parallel with the conductivity of the tube. We have not considered the complications, if any, that might result when attempting to measure the density of a highly conducting liquid (e.g., a liquid metal). The results in Sec. V below indicate that no problems were encountered using a liquid as conducting as distilled water.

IV. OSCILLATING TUBE ASSEMBLY

Figures 1 and 2 display one of our simplest and most successful densimeters. Except where otherwise noted, the details in this manuscript apply to this instrument, although we have made more than half a dozen others. We now describe, in turn, the oscillating tube, the body of the densimeter that supports the tube and the magnet, the electrical connections to the tube, and the magnet.

A. Oscillating tube

Oscillating tubes were formed from several alloys and tested. Satisfactory performance was obtained with stainless-steel 316, Inconel 600, and Hastelloy C with our test liquids, water, and toluene. In general, the nickel-based alloys showed less color change resulting from baking in air at 675 K in our furnace.

In order to keep the densimeter compact, the outside diameter of the tube was chosen to be 1.65 mm (0.062 in). (In Fig. 2, the size of the densimeter is indicated by the centimeter scale shown in the photograph.) With the outside diameter fixed, the sensitivity of the oscillator's resonant frequency to density decreases as the thickness of the tube's wall is increased. Thus, we chose the wall's thickness to be 0.25 mm (0.010″), a value that is near the minimum consistent with our pressure requirement. Based on handbook values for the mechanical properties, the Hastelloy C tube had a tensile strength of 700 MPa at room temperature. However, we did not test the tube at pressures above 20 MPa (corresponding to a tensile stress of 140 MPa) at 575 K.

From a longer piece of tubing received from the manufacturer, we cut a length of 1.2 m. The center portion of this piece was bent over a jig into a half circle slowly to avoid kinking the tube.

The ends of the oscillating tube extended 55 cm without joints from the densimeter through a small hole in the furnace into the laboratory where compression fittings were used to fasten it to a manifold that supplied the test liquids and applied pressure to them.

B. Densimeter body

The body of the densimeter must firmly support the tube and the magnet and is must electrically insulate the two ends of the tube from each other. As shown in Figs. 1 and 2, the body of the densimeter was comprised of six metal parts that were machined out of plate stock. Both stainless-steel 304 and Inconel 600 plates were successfully used. The key part is a chair-shaped plate 6.35 mm thick that is at the top of Fig. 1 and the front of Fig. 2.

Both legs of the oscillating tube near the U-bend were clamped into grooves machined into metal plates. One leg was clamped between the chair-shaped plate and a metal block; the other leg was clamped between two metal blocks. The base plate and the three blocks were bolted together with two 10–32 stainless-steel threaded rods and nuts. There was no indication that differential thermal expansion between the bolts and the plates caused difficulties. The threaded rods were longer than necessary to hold the blocks together. The extra lengths had nuts and washers on them so that they could be used as binding posts for connecting the electrical leads to the oscillating tube.

The magnet was clamped between two metal blocks with two 10–32 stainless-steel bolts. These bolts were screwed into holes that had been tapped through the larger of the two blocks. The larger block itself was bolted to the chair-shaped plate with two more stainless-steel bolts, nuts, and washers.

The tube of the densimeter shown in Figs. 1 and 2 was forced to oscillate in the field of the permanent magnet by passing an alternating electrical current through the tube itself. This arrangement requires that the metal plates that clamp the two ends of the tube be electrically insulated from each other. The insulation was achieved by separating the two sets of the grooved plates with mica sheets about 0.5 mm thick. We tested much thicker sapphire and machinable ceramic slabs as insulators, however they cracked after a few thermal cycles. The thin mica sheets have survived numerous thermal cycles without apparent damage.

C. Electrical connections to the oscillating tube

As shown in Fig. 3, the oscillating tube is connected to four electrical leads. If a low impedance detector is used, the signal wires should have a low resistance in addition to satisfactory heat resistance. We used commercially available nickel-plated copper wires (AWG size 22) insulated with inorganic sleeving and braid rated to 1090 K by the manufacturer. We also found that solid gold wires approximately 0.6 mm in diameter insulated with fiberglass sleeving of the proper temperature rating were satisfactory.

The portion of the U-shaped oscillating tube between the clamping points was approximately 100 mm (4 in.) long and it had a resistance of 0.07 Ω at ambient temperature. To avoid loss of signal, the electrical resistance of the mica sheets at 575 K must be much larger than 0.07 Ω, a requirement that was easily met.

The electrical resistance of the entire densimeter tube extending from the manifold at ambient temperature into the furnace and back to the manifold was approximately 0.7 Ω. This was sufficiently large that the electrical short circuit between the ends of the tube *via* the metal manifold did not materially reduce the signal available at the detector.

D. Magnet

The strength of the magnetic field is an important design parameter because the force driving the tube is proportional to the square of the field. The magnet was made of Alnico V with a Curie temperature of about 990 K; thus, it was suitable for operation at the temperatures encountered here.

As shown in Figs. 1 and 2, the oscillating tube passes through a narrow gap between the poles of a C-shaped permanent magnet. This shape of magnet yields a high magnetic field for a given amount of magnetic material and avoids the use of pole pieces. (Pole pieces are another potential source of mechanical instability because they might move in response to the variations in the magnetic force on them accompanying the motion of the tube.) The magnetic field in the gap was about 0.35 T and the magnet weighed only 54 g. The outer diameter of the magnet was 38.1 mm (1.5 in.); the gap was 2.9 mm (0.114 in.) wide and 12.7 mm (0.5 in.) thick.

V. EXPERIMENTAL PROCEDURES AND RESULTS

A. Initial heat treatment

Initially, we expected the resonance frequencies of the densimeters to decrease as they aged in the furnace. Presumably, progressive oxidation would increase the effective mass of the tubes and annealing would decrease their stiffnesses. To our surprise, we observed that the resonance frequencies of all of our densimeters drifted upward when they were first heated following assembly.

The drift rate was higher at higher temperatures. Typically, the initial drift rate was 1,000 ppm (parts per million) per day when a densimeter was first brought to 670 K. However, there were cases when the initial drift rate was 3,000 ppm per day. In one case, an initial increase of the resonance frequencies occurred even when the U-shaped tube had been annealed in vacuum at 1400 K prior to the assembly. When a densimeter was kept at a constant high temperature, the drift rate gradually decreased over a period of days; however, it did not vanish. For example, baking one densimeter at 670 K for three days reduced the drift rate at 570 K to a few parts per million per day and the drift rate at still lower temperatures became too small to be detected in less than a day.

These observations led us to adopt a heat treatment regimen for each newly assembled densimeter before putting it into service. Each densimeter was cycled between 570 and 670 K. The temperature was changed once each day and the $n=3$ resonance frequency was continuously monitored. The heat treatment was terminated when the drift rate nearly vanished at 570 K. Typically, this process lasted about five days. We did not study the behavior of densimeters above 670 K because of the accelerated oxidation of the tubes at these higher temperatures.

We also monitored the quality factors Q of newly assembled densimeters. When first assembled, a typical value of Q is 2000 at room temperature. Following heat treatment the Q was a factor of 3 to 5 higher.

B. Calibration and test measurements

The two calibration functions, $k(T)$ and $v(T,p)$, in Eq. (4) were determined from frequency measurements $f_v(T)$ and $f_w(T,p)$, that is measurements when the densimeter was

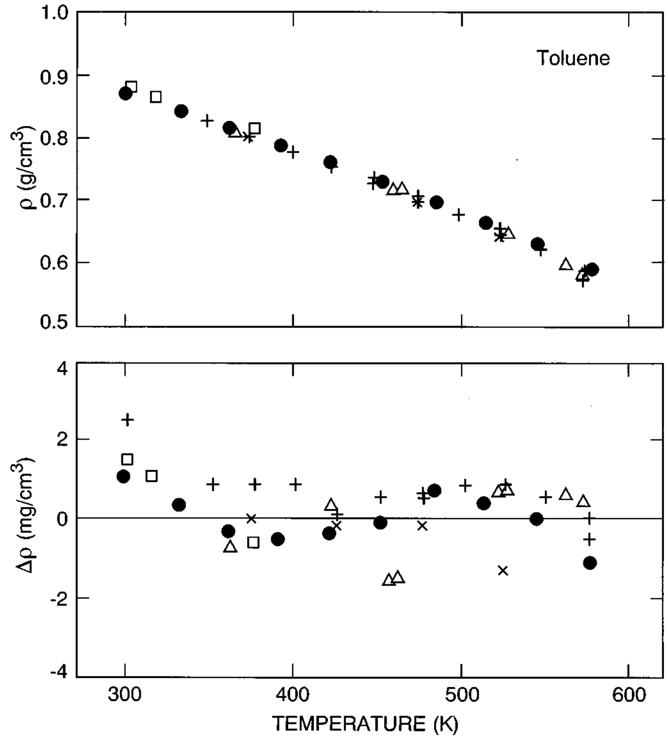

FIG. 6. Top: The density of the toluene as a function of temperature at a pressure of 13.8 MPa as determined from the resonance frequencies. Bottom: Deviations of the measured densities from the correlation of Goodwin.[9] Key: (●) present measurements; (+) Akhundov and Abdullaev,[10] (×) Kragas *et al.*,[11] (□) Kashiwagi *et al.*,[12] and (△) Straty *et al.*[13] From Refs. 10–13, only the data in the pressure range 10–15 MPa are plotted. The rms deviation of the present measurements from the correlation is 0.61 mg/cm^3.

evacuated and when it was filled with water at various temperatures and pressures. The function $k(T)$ was represented by a polynomial function of the temperature

$$k(T)=f_v^2(T)=F_0+F_1T+F_2T^2+F_3T^3+F_4T^4 \tag{12}$$

and the function $v(T,p)$ was determined from

$$\begin{aligned} v(T,p)&=[f_v^2(T)/f_w^2(T,p)-1]/\rho_w(T,p)\\ &=(V_0+V_1T+V_2T^2+V_3T^3)(1+V_pp), \end{aligned} \tag{13}$$

where $\rho_w(T,p)$ was obtained from the *Steam Tables*[8] at the same values of T and p that were used in measuring $f_w^2(T,p)$. The water had been degassed by boiling prior to being loaded (under vacuum) into the densimeter. The calibration data spanned the range 298 K$<T<$575 K at the pressure $p=13.8$ MPa. The calibration functions were inserted in Eq. (4) to compute the density of test fluids from measurements of the resonance frequency.

Toluene was used as a test liquid to assess the performance of the densimeter. This particular densimeter had stainless-steel 316 for the tube. The toluene data were acquired at ten temperatures in the range 298 to 575 K while the toluene was pressurized with helium to 13.8 MPa.

The results for toluene are shown in Fig. 6, where they are compared to the correlation of Goodwin.[9] Figure 6 also displays the data from the literature in the range 10 MPa $<p<$15 MPa. These data are a subset of those that Goodwin

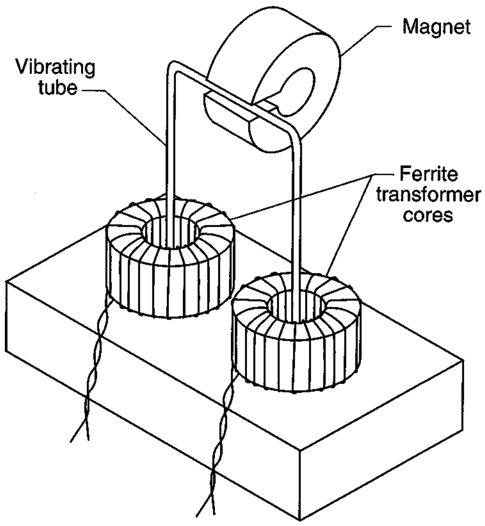

FIG. 7. Schematic drawing of an oscillating tube with inductive coupling. For clarity, only the essential parts are drawn.

used to construct his correlation. None of them were acquired with an oscillating tube densimeter. The rms deviation of our data from the correlation is 0.61 mg/cm^3, which is slightly less than 0.1% of the density of toluene under these conditions. The deviation plot (bottom of Fig. 6) suggests that the present data are in slightly better agreement with the data underlying the correlation than with the correlation itself. This is not surprising because the correlation spans a much wider range of conditions than do the present data. The scatter in the present data is less than the scatter in some of the data sets from the literature.

To assess the stability of the densimeter we compared two series of measurements of $f_v^2(T)$ for $298<T<575$ K obtained ten days apart. The deviation of the second series from a function of temperature representing the first series was a smooth function of the temperature with an rms deviation equivalent to a density change of 0.25 mg/cm^3.

VI. MAGNETIC COUPLING FOR EXCITATION AND DETECTION

In some circumstances, it may be desirable to improve the mechanical stability of the densimeter by not having a layer of insulation between the two legs of the oscillating C-shaped tube and by not bolting parts together. This can be achieved by inductively coupling to the tube. We have demonstrated this concept at ambient temperature.

As illustrated in Fig. 7, the demonstration setup used a U-shaped tube that had been threaded through two ferrite torroids and then through two clearance holes that had been drilled through a single stainless-steel plate. The tube was brazed into the holes forming an electrically closed stainless-steel loop with a resistance of approximately 0.07 Ω. (Although technically more demanding, the tube could have been welded to the plate allowing operation at temperatures where the brazing alloy becomes soft.) In effect, the brazed stainless-steel loop functioned as a one-turn shorted winding for two independent transformers. The ferrite torroids were the cores of the transformers. One transformer coupled the driving current into the loop; for this transformer, the loop was the secondary coil. The other transformer was used to detect the current circulating in the loop. (At resonance, this current is reduced by the induced emf.) For this detecting transformer, the loop was the primary coil. As sketched in Fig. 7, the additional windings that had been placed around the ferrite cores had multiple turns to help match the low impedance of the loop to the higher impedances of the oscillator and detector. When the frequency of the oscillator was scanned across the densimeter's resonances, we measured a frequency response similar to that shown in Fig. 5, except that the detected voltage decreased at resonance.

In our realization of this concept, the mica insulation in the structure of the densimeter was replaced by the insulation on the windings on the ferrite cores. In this new location the insulation is not subjected to strong compressive forces that were applied to the mica. Indeed, the transformer cores could have been independently supported and then the aging of their mechanical properties would not affect the resonance frequencies. If the transformers degrade with age, the sensitivity of the circuits that drive and detect the oscillations would be changed; however, the resonance frequencies from which the density is deduced would not.

ACKNOWLEDGMENT

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